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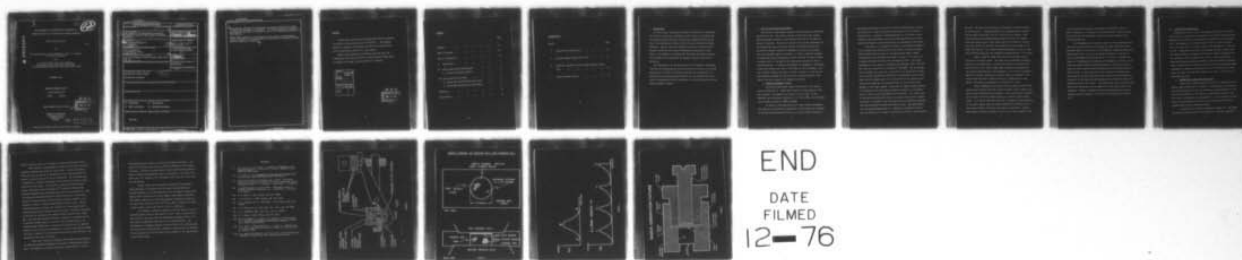SEP 76 G PORTER, C J TREDWELL, H D FAIR   DA-ERO-75-G-058

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NEW TECHNIQUES FOR INVESTIGATING PROPERTIES OF

ENERGETIC SOLIDS AT HIGH STATIC AND DYNAMIC PRESSURES

Final Technical Report

by

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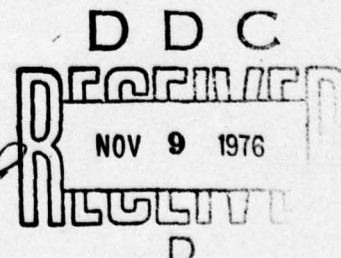(Visiting Research Scientist, Royal Institution, 1975)

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Stress waves induced by a Q-switched ruby laser pulse are investigated, and the feasibility of using picosecond light pulses to generate far larger pressure gradients is examined.

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investigated, and the feasibility of using picosecond light pulses
to generate far larger pressure gradients is examined.

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CONTENTS

ILLUSTRATIONS

I. INTRODUCTION:

In recent years, considerable progress has been made in determining the electronic structure of energetic solids (1) and in elucidating the relationship between electronic structure and chemical instability (2). Most of the experimental research to date has been performed at ambient pressures. However, since fast chemical reactions in a broad class of energetic solids are initiated by subjecting the materials to shock waves, it is important to determine the electronic and lattice structure and processes at the high static and dynamic pressures experienced in actual use.

The objectives of the current work are to (1) develop a microscopic technique for determining the optical properties of solids at static high pressures up to 50 Kbar, (2) develop techniques for the generation of shock waves in solids using pulsed lasers, and (3) determine the feasibility of measuring the optical properties of a solid subjected to a high pressure gradient (shock).

II. STATIC HIGH PRESSURE MEASUREMENTS:

This portion of the program utilized a newly designed and fabricated diamond anvil cell. This unique cell was constructed by Drs. Stanley Block and Gaspar Piermarini at the Materials Institute of the National Bureau of Standards. This type of cell has been utilized extensively for the visual observation of pressure effects on materials but had been limited by a general inability to determine the value of the pressure with any reasonable degree of certainty. A rapid, convenient technique for precision pressure measurement has been developed (3) which utilizes a pressure shift in the sharp R-line fluorescence spectrum of ruby or similar materials. In addition, new designs for alignment of the diamond anvils have extended the range to 200 kilobar at room temperature. The cell utilized in these experiments is shown in Figure 1. This cell has been specially designed and constructed with hollowed channels in the body of the cell to permit low temperature operation.

A. PRESSURE MEASUREMENT SYSTEM:

The pressure measurement system utilized here is the same as given in reference (3). Small single crystals of Ruby (Al_2O_3:Cr 0.5%) were utilized. The pressure coefficient for Ruby is $\frac{d\lambda}{d\rho} = + 0.365$ Å/kilobar and the spectral linewidth is 7.5Å (full width at half maximum) for the doublet R-lines at 6928Å and 6942Å.

To obtain a truly hydrostatic (zero shear stress) environment, the sample to be studied and a small piece of Ruby crystal are contained in the same encapsulating fluid used as a pressure transmitting medium.

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Maintaining hydrostatic conditions in systems at high pressures requires the selection of a well-characterized fluid which will not solidify or become viscous. A mixture of methanol:ethanol (4:1 by volume) is used throughout these experiments since it has been demonstrated (4) to be a superior hydrostatic transmitting fluid up to approximately 100 kilobar.

The sample is contained in a chamber as illustrated in Figure 2. A low strength metal (Inconel 600) 10 mils thick is used as the gasket material which forms the walls of the high pressure sample chamber. Conventionally, the extremely small sample is taken to be a grain or so of powder surrounded by the pressure transmitting fluid. For optical measurements in the regions of extremely high absorption ($10^5 - 10^6$ cm^{-1}) thin films are required with a thickness of 500-5000Å. The top diamond-anvil is masked and coated with a thin film of the appropriate material. One of the major experimental difficulties in performing optical experiments with the diamond anvil cell is associated with the optical precision and care which must be employed because of the extremely small diameter of the sample chamber. Recall that the sample chamber consists of a cylinder in the steel gasket formed by drilling a 0.3 mm diameter hole in a piece of steel approximately 0.07 mm thick. The central part of the system utilized here consists of a sturdy, high quality research microscope. The movable stage was removed but the adjustable condensing lens and polarizing accessories were kept in place. The high pressure cell was mounted independently on a rugged X-Y-Z micrometer system so that the position of the sample volume could be precisely controlled and

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adjusted. The focus and positioning of the sample is obtained entirely by the XYZ device. The high pressure cell is approximately 3 cm thick and the sample volume lies 1 1/2 to 2 cm from the top surface. This requires special long working distance objectives since the standard microscope objective has a short focal length so the width of the pressure cell prevents the objective from being placed close enough to the sample for focusing. For a sample volume with diameter 0.3 mm, at least three different objectives are required with magnifications 3, 10 and 20X.

Figure 2 shows the image observed with a small Ruby crystal enclosed in the sample volume. The light may be transmitted through an eye piece to a monochromator. The ruby fluorescence is extremely efficient and with a low f-number (high light transmission) monochromator, red-sensitive photomultiplier and picoammeter, it is entirely possible to detect the red Ruby luminescence and hence measure the pressure in the sample volume even with a sample as small as a single grain of ruby powder a few tenths or hundredths of a millimeter on a side.

Using a diaphragm at the proper position, specific areas of the volume may be isolated optically so that particular areas of single crystals or thin film samples may be studied. This is particularly important in the study of thin film samples which are not purely single crystal films. It is general experience with thin film samples of many materials to form small regions which are single crystalline, but which are oriented along different directions with respect to each other. Optical measurements performed on samples of this type are then averages of the

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various orientations leading to broadening of anisotropic lines, etc.
Using a diaphragm, it is possible to localize specific crystalline areas
and thus observe the properties of single crystalline regions of the
polycrystalline film.

The diaphragm must be placed at the focus of the microscope and
there must be some provision for observing the image (preferably under
polarized light) at the position of the diaphragm. Using objectives with
magnifications of 3, 10, 20, it was found satisfactory to fabricate a
series of diaphragms with openings from 0.2 to 0.9 mm. To observe the
image at the diaphragm and to position the diaphragm at the focus, an eye
piece was utilized which was normally designed to contain cross hairs.
These are easily removed and metal diaphragms may be quickly inserted in
place of them. It was observed that this technique also enhances the
sensitivity of the ruby luminescence measurement since the diaphragm can
exclude all of the light from the monochromator and detector except that
emanating from the ruby crystal. The high pressure cell position is then
moved until various parts of the sample are in the optical path of the
diaphragm. A number of inorganic semiconducting materials were inves-
tigated using this technique. The results are summarized in reference
(13).

III. LASER-INDUCED STRESS WAVES:

Investigations (5, 6, 7, 8) of the plasma produced when Q-switched laser pulses are absorbed on solid material surfaces have shown that high amplitude stress pulses or shock waves are generated in the target material. The magnitude of the stress using a nominal 2 Joule Q-switched laser is on the order of 1 kilobar (1kbar = 986 atmospheres = 14,494 psi) or less. Anderholm (9) found that by confining the laser induced "blow-off" plasma between a transparent solid and the target, the magnitude of the stress wave could be enhanced by at least an order of magnitude. Using the Anderholm confinement technique, Yang (10) studied the dependence on target material, thickness and laser fluence and described simple techniques for obtaining stress waves of from 0 to 20 kbars with stress pulse rise times and half-widths (FWHM) on the order of 20 nsec from a Q-switched Ruby laser.

A. INTERACTION OF SHOCK WAVES WITH SOLIDS:

A shock wave or high intensity stress wave is produced by an extremely rapid acceleration of a surface. Traditionally, (11) the means for this acceleration has been the detonation of high explosives in contact with the material or the impact of a high-speed projectile fired from a gun or accelerated by high explosives. In the technique utilized here, the stress waves are the result of the work done by the laser-induced plasma on the confining materials.

A stress wave in a solid is exhibited in Figure 3a. The region AB in which the stress rises to its maximum is called the compression

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phase of the wave; the region BC is called the rarefaction phase. The velocity of propagation associated with the peak of an intense stress wave, where the material is at a high pressure and density, may be greater than the velocity at the extreme front of the wave where the pressure and density are close to the initial undisturbed state. Thus, the profile of the wave changes in time to become an almost discontinuous jump in pressure and density at the wave front followed by a gradual fall (see Figure 3b). This type of stress wave is called a shock. As transmission of the wave continues through the solid, losses gradually reduce the peak pressure and the shock wave ultimately degenerates into an ordinary sound wave.

When the shock wave reaches a free surface or boundary between two media, it undergoes reflection or refraction depending essentially on the relative densities and sound velocities of the materials. For example, a shock wave travelling through a dense medium is totally reflected at the boundary or solid-vacuum interface. The reflected wave is led by a rarefaction front. At the plane of interaction of the reflected wave and the rarefaction phase of the initial stress wave, a tension is induced. If the tension exceeds the dynamic tensile strength of the material, the material undergoes fracture or spallation.

B. STRESS WAVE GENERATION AND MEASUREMENT:

Stress waves were generated by irradiating a thin metal film confined between glass and metal substrates with a pulsed Ruby laser (see Figure 4). The intensity and duration of the stress waves were determined

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by measuring the piezoelectric current induced in x-cut quartz single crystals.

A Ruby Laser was Q-switched using vanadyl phthalocyanine in nitrobenzene and focused onto a thin film of aluminium as shown in Figure (4). A small portion of the laser pulse was passed through a fast photo-diode and the output recorded on a Tektronix Model 7904 fast rise time oscilloscope. The laser output was maintained at 1.8 Joules and monitored on a calorimeter. The rise time to the laser peak power and the half width (FWHM) are approximately 20 nsec. Irradiation of a 1 micron thick aluminium film with the 1.8 Joule pulse produces a shock wave of approximately 10 kbar peak pressure with a rise time of approximately 20 nsec and half width of 25 nsec (FWHM).

A major achievement in Explosives Research would be to obtain detailed information about the microscopic chemical and physical processes occurring in the shock front of an explosive solid. An obvious initial experiment is to perform time-resolved optical spectroscopy in an explosive material as a shock wave passes through it. However, one of the major experimental obstacles to such a study is synchronization of the time-resolved experiment with the generation or passage of a shock wave through the material.

This portion of the research program investigated the possibility of exploiting advances in mode-locking solid state lasers and in selecting individual pulses from a mode-locked train to obtain from a single laser shot, the optical energy for generation of a shock wave and the temporally

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separated optical pulses for performance of time-resolved spectroscopy.

Mode-locking is accomplished by inserting an ultra-fast intensity dependent bleachable dye inside the laser cavity. A pulse oscillating within the laser cavity will be amplified only if it has sufficient intensity to cause the dye to bleach to transparency. The dye should have a relaxation or recovery time shorter than the time for a pulse to make a round trip in the cavity. The result is a series of extremely narrow pulses, each pulse a few picoseconds (10^{-12} seconds) in duration and separated temporally from contiguous pulses by the time required for light to traverse the distance between the Fabry-Perot mirrors of the cavity. Using an electronic pulser and electro-optic selection elements, it is possible to extract a single picosecond pulse from the train. This is achieved by placing a Pockels cell (energized by a spark gap) between crossed polarizers down stream from the oscillator. Initially, each pulse in the train is rejected by the second polarizer and sent into a spark gap. The spark gap breakdown threshold is adjusted to correspond to one of the most intense pulses in the train. When breakdown occurs, the resulting high voltage pulse is applied to the Pockels cell for approximately 3-5 nsec so that the polarization of the next pulse passing through the cell is rotated 90° and transmitted through the second polarizer. The remainder of the train is rejected.

There are a variety of techniques which enable optical pulses to be temporally separated depending on the needs of the particular experiment. For example, an extremely simple but elegant method is to use a

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stepped-optical delay echelon to provide the temporal separation. The echelon is fabricated from a stack of ordinary microscope slides (glass or quartz). Identically stepped delays are generated between portions of light traveling through adjacent parts of the echelon. For slides 1 mm thick and at 45° incidence, the intersegment delay times are on the order of 3 picoseconds.

Finally, models of the processes occurring in laser-induced shock generation (12) indicate that the energy loss mechanisms are strongly dependent on the laser deposition time and predict significantly higher stress pressures as the laser temporal pulse width is decreased. For example, keeping all of the laser parameters constant but reducing the laser pulse width from 10^{-6} to 10^{-12} sec leads to predicted increases in peak pressure from on the order of 1 kbar to 1,000 kbar.

Our attempts to measure a significant pressure from an extracted picosecond pulse have been unsuccessful to date. This is clearly due to the low flux of the pulse from our experimental arrangement since it is many orders of magnitude lower than for the Q-switched pulse. Our measurements do establish, however, the feasibility of the technique and, with an optically amplified pulse, sufficient intensity will be available for the generation of the shock wave and performance of the time-resolved optical spectroscopy.

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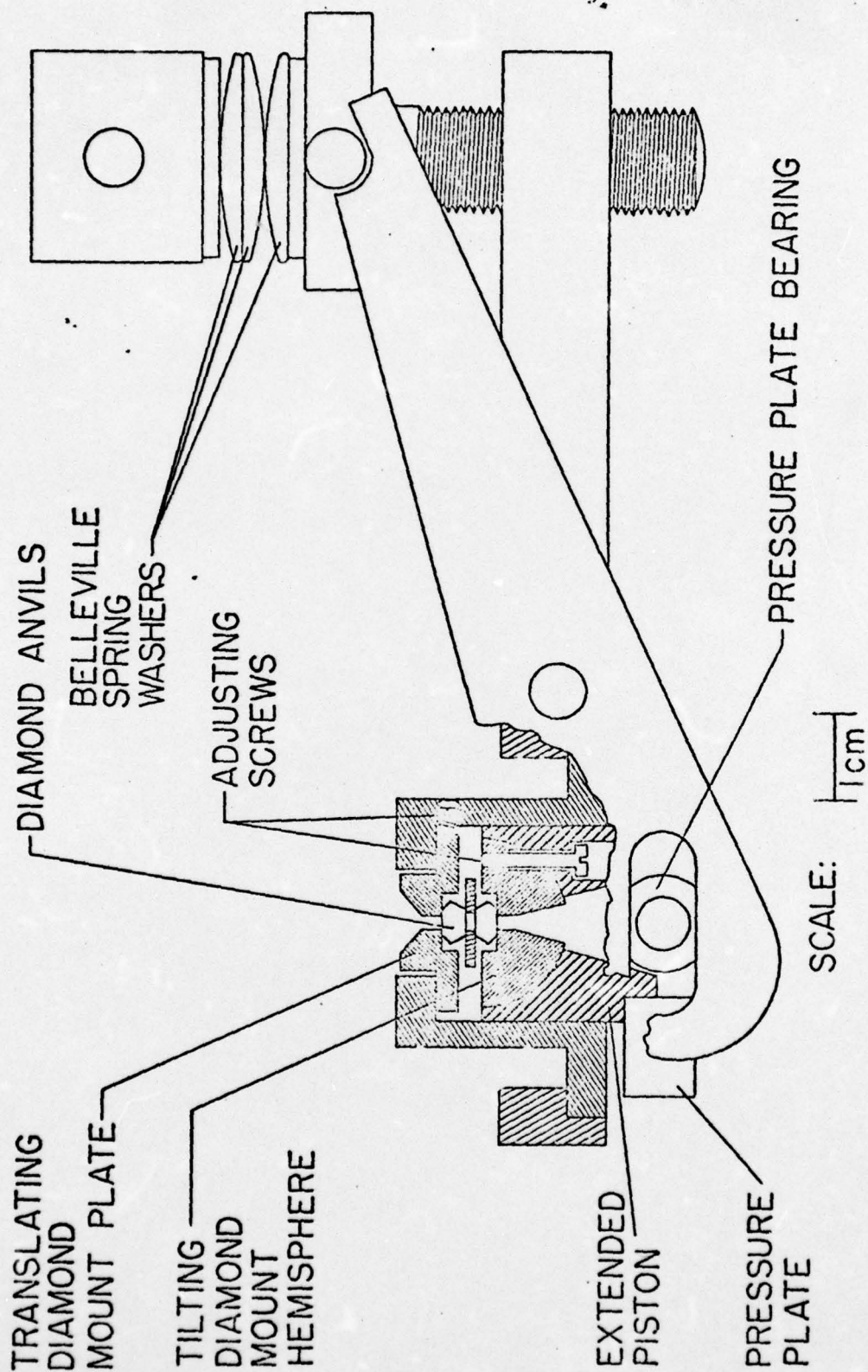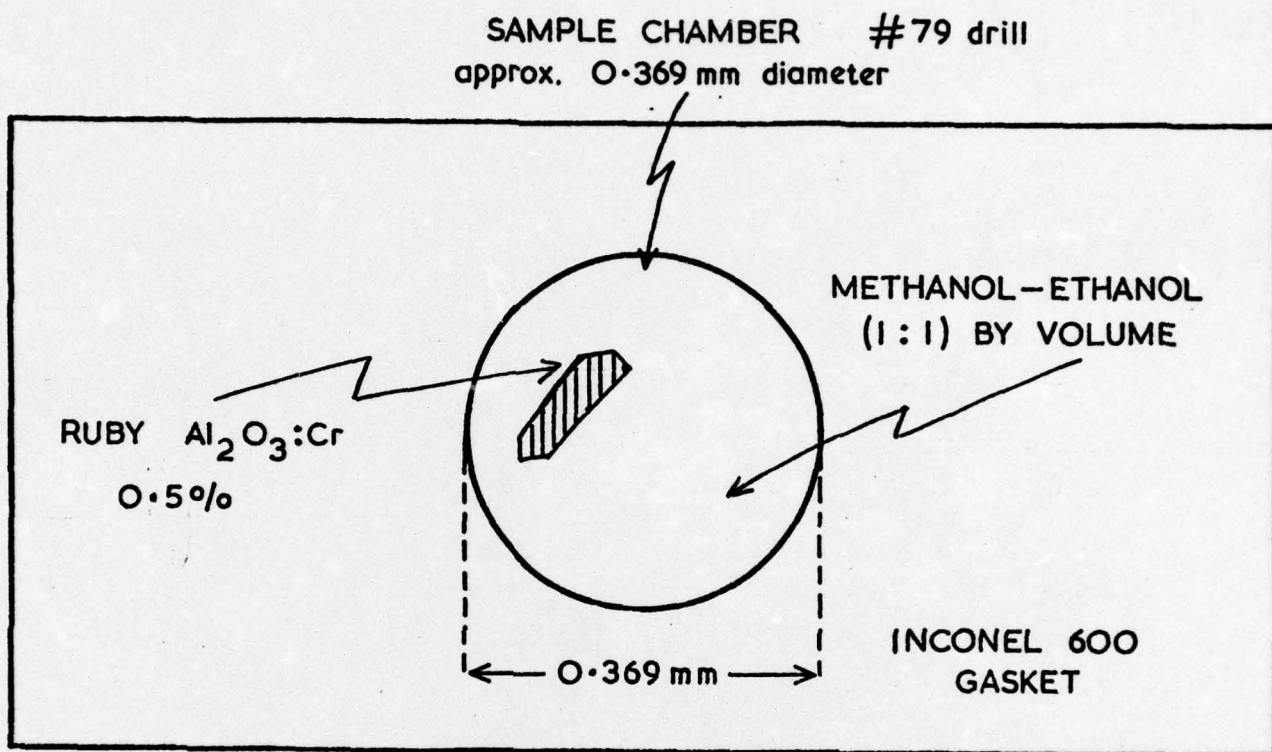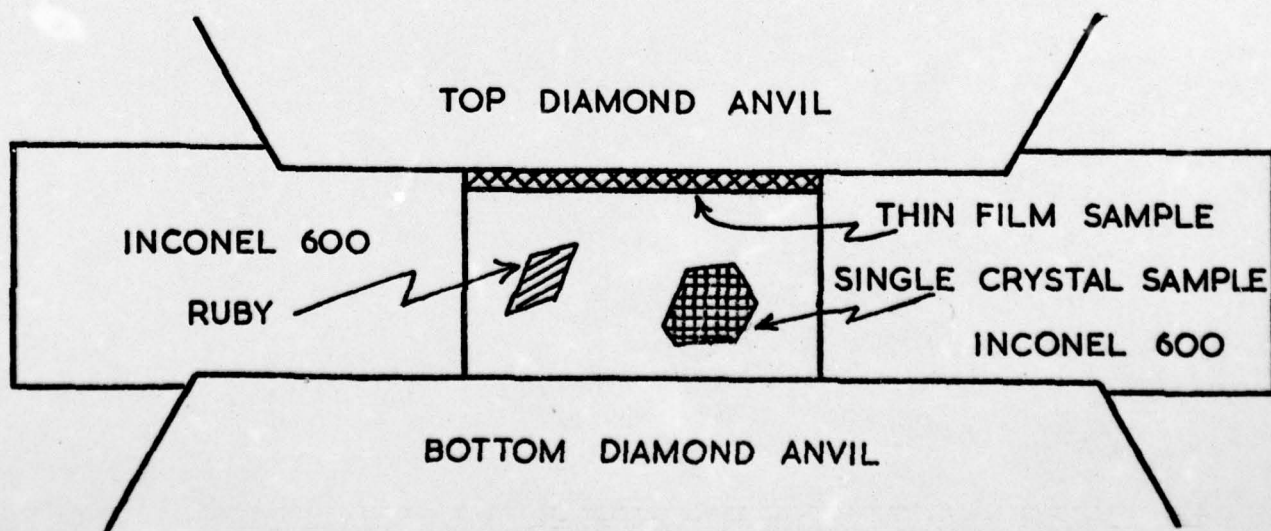
DIAMOND ANVILS

BELLEVILLE
SPRING
WASHERS

ADJUSTING
SCREWS

TRANSLATING
DIAMOND
MOUNT PLATE

TILTING
DIAMOND
MOUNT
HEMISPHERE

EXTENDED
PISTON

PRESSURE
PLATE

PRESSURE PLATE BEARING

SCALE: |1cm|

FIGURE 1

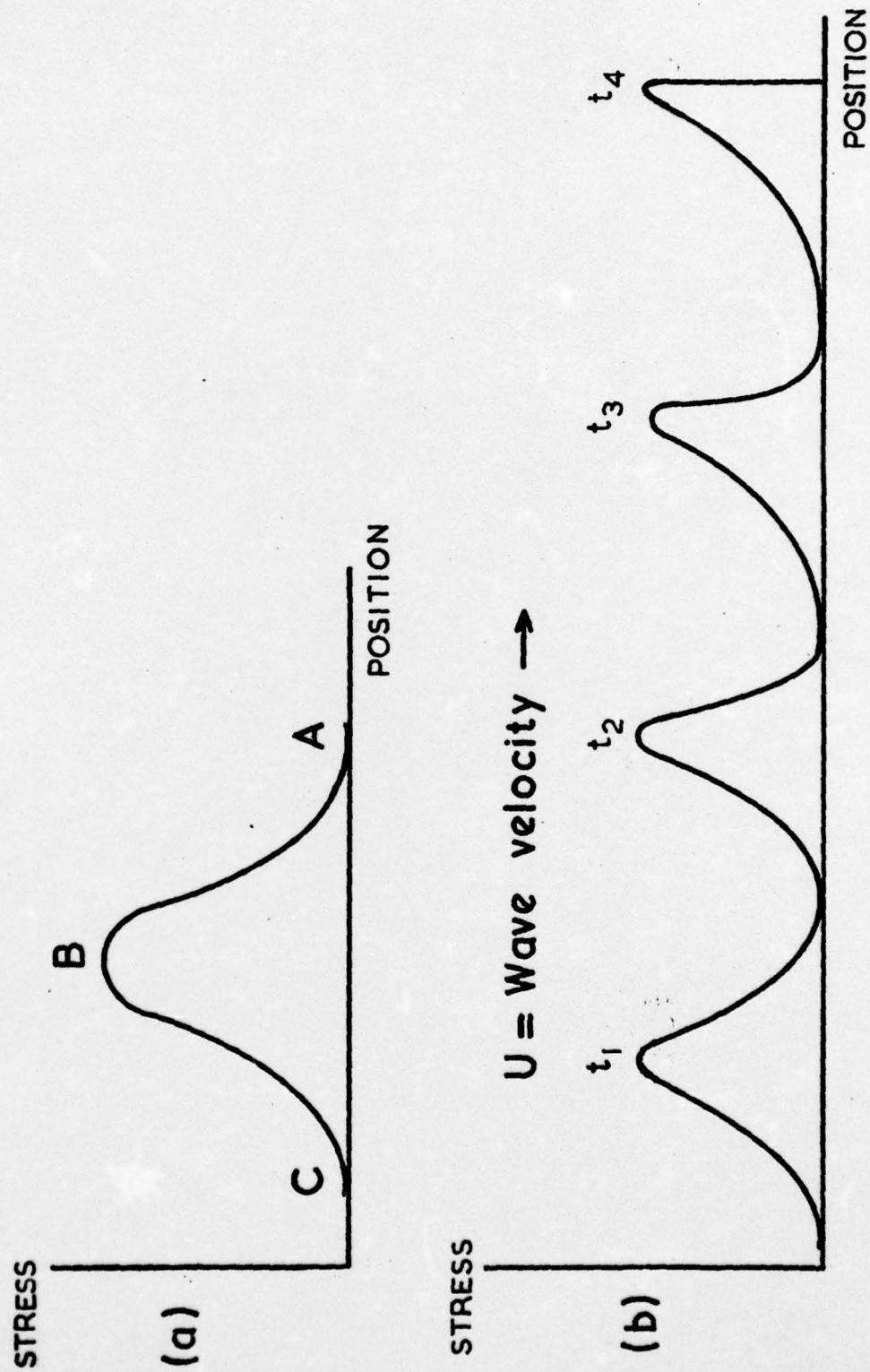# SAMPLE CHAMBER OF DIAMOND ANVIL HIGH PRESSURE CELL

TOP VIEW

SIDE VIEW

FIGURE 2

STRESS

POSITION

B

A

C

(a)

STRESS

U = Wave velocity →

t_1 t_2 t_3 t_4

POSITION

(b)

FIGURE 3

SHOCK GENERATION FIXTURE

NYLON PLUG

STAINLESS STEEL SCREW ELECTRODE +

INDIUM FOIL

QUARTZ CRYSTAL

BRASS FOIL

STAINLESS STEEL BODY

GLASS WINDOW

LASER

THIN FILM

FIGURE 4